

Quasi-classical cyclotron resonance of Dirac fermions in highly doped graphene

A. M. Witowski,¹ M. Orlita,^{2,3,4,*} R. Stępniewski,¹ A. Wymolek,¹
J. M. Baranowski,^{1,5} W. Strupiński,⁵ C. Faugeras,² G. Martinez,² and M. Potemski²

¹*Institute of Experimental Physics, University of Warsaw, Hoża 69, 00-681 Warsaw, Poland*

²*Laboratoire National des Champs Magnétiques Intenses,*

CNRS-UJF-UPS-INSA, 25, avenue des Martyrs, 38042 Grenoble, France

³*Institute of Physics, Charles University, Ke Karlovu 5, 121 16 Praha 2, Czech Republic*

⁴*Institute of Physics, v.v.i., ASCR, Cukrovarnická 10, CZ-162 53 Praha 6, Czech Republic*

⁵*Institute of Electronic Materials Technology, Wolczynska 133, 01-919 Warsaw, Poland*

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Cyclotron resonance in highly doped graphene has been explored using infrared magneto-transmission. Contrary to previous work, which only focused on the magneto-optical properties of graphene in the quantum regime, here we study the quasi-classical response of this system. We show that it has a character of classical cyclotron resonance, with an energy which is linear in the applied magnetic field and with an effective cyclotron mass defined by the position of the Fermi level $m = E_F/v_F^2$.

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I. INTRODUCTION

The cyclotron motion of charge carriers with a well defined effective mass m and the related resonant absorption of light at the cyclotron frequency $\omega_c = |e|B/m$ are basic physical phenomena representative of the magneto-optical response of conventional condensed-matter systems. Well-established and often applied, cyclotron resonance (CR) became through the years a common technique routinely used in solid-state physics, inherently connected with the effective mass of elementary electronic excitations.¹

Recently fabricated graphene^{2,3} immediately became a challenge for CR measurements and implied necessity to comprehend the phenomenon of cyclotron resonance in a system with massless charge carriers.^{4,5} Special character of CR in graphene can be exemplified on the basis of a simple quasi-classical approach.⁶⁻⁹ If we solve the classical equation of motion for a charged particle with an energy ε which depends linearly on the momentum p , i.e. $\varepsilon = \pm v_F|p|$, we find out that the precession-like (cyclotron) motion remains characteristic also for these Dirac-like particles, nevertheless, with a cyclotron frequency modified to $\omega_c = |e|B/(|\varepsilon|/v_F^2)$. Hence, the classical cyclotron motion and consequently also the cyclotron resonance of massless Dirac fermions scale linearly with the applied magnetic field B , similar to conventional massive particles, but they depend on the particle energy, which appears in a form of an energy-dependent cyclotron mass defined by the Einstein relation $|\varepsilon| = mv_F^2$.

Surprisingly, this classical, i.e. linear with the magnetic field, cyclotron resonance has not been clearly observed in graphene so far. Instead, the optical response of graphene has been experimentally explored¹⁰⁻¹³ only in the quantum regime, when all detected transitions correspond to excitations between well-separated Landau levels (LLs). These measurements provided us with a unique insight into the LL spectrum of graphene and di-

rectly visualized its intriguing \sqrt{B} -scaling, but no information about the quasi-classical limit could be extracted. Consequently, it remains a challenge to demonstrate experimentally the quasi-classical cyclotron resonance in graphene, or interestingly, to study the crossover between this classical (linear in B) and fully quantum-mechanical (linear in \sqrt{B}) magneto-optical response of graphene – the crossover, which is not present in a system of massive particles, where the response always remains linear in the applied magnetic field.

Here we report on magneto-optical measurements on highly-doped epitaxial graphene grown on the silicon-terminated surface of SiC. We identify the cyclotron-resonance-like response related to the massless Dirac fermions in the vicinity of the Fermi level, which evolves nearly linearly with the applied magnetic field, as expected from (quasi-)classical considerations. These results are compared to the currently well-understood response of multilayer epitaxial graphene prepared on the carbon-terminated surface of SiC, where majority of (rotationally stacked) graphene sheets is nearly undoped and where the fully quantum-mechanical treatment is appropriate for its description.

II. SAMPLE PREPARATION AND EXPERIMENTAL DETAILS

In our study, we present data taken on two particular samples of graphene prepared by thermal decomposition on the surface of semi-insulating silicon carbide.³ Both specimen have been grown at 1600°C in Ar atmosphere using a commercially available horizontal chemical vapor deposition hot-wall reactor (Epigress V508), inductively heated by a RF generator.

The sample denoted as A in our study, grown on the C-terminated surface of silicon carbide (4H-SiC[000 $\bar{1}$]), represents a standard multilayer epitaxial graphene (MEG)

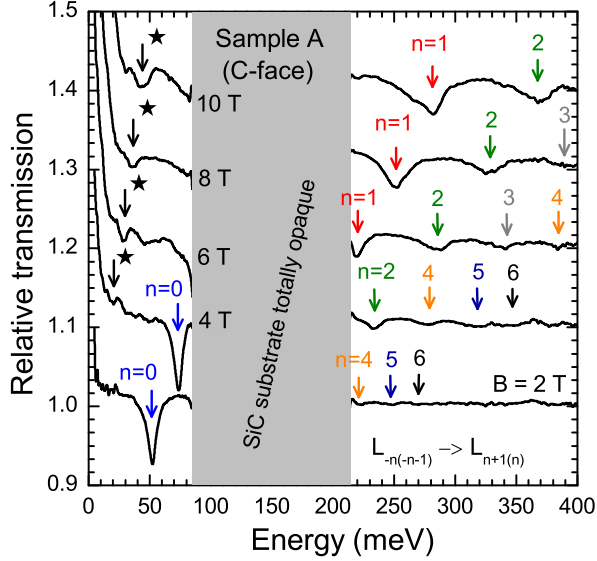


FIG. 1: (color online) Relative transmission spectra of the sample A (MEG on C-face of SiC) for selected values of the magnetic field. For clarity, successive spectra are shifted vertically by 0.1. Individual transitions discussed in this paper are denoted by vertical arrows.

specimen. The majority of sheets in this structure is practically undoped (residual p -like doping expected at the level of 10^9 cm^{-2} , see Refs. 13 and 14) with the electronic band structure similar if not identical to an isolated graphene monolayer, provided by the characteristic rotational stacking of layers.¹⁵ This fact is reflected in the micro-Raman spectra taken on this sample, which revealed dominantly single-component (Lorentzian-like) 2D band,^{16,17} nevertheless, some Bernal-stacked residuals have been observed on selected locations as well.

The sample B has been prepared on the Si-terminated surface of SiC (4H-SiC[0001]) and also characterized in the micro-Raman experiment. Similarly to the previous sample, the dominantly observed single-component 2D band serves as an indication for linearity of electronic bands. It points towards the presence of a single graphene sheet (in addition to the non-graphene-like buffer layer¹⁸), as in contrast to MEG on the C-face, the few-layer graphene stacks on the Si-face exhibit Bernal and not rotational ordering.¹⁹ In addition, such monolayers are typically rather highly doped by the charge transfer from the substrate.^{20,21}

To measure the (non-polarized) infrared transmittance of our samples, we used the radiation of a global, which was analyzed by a Fourier transform spectrometer and delivered to the sample via light-pipe optics. The light was then detected by a composite bolometer kept at $T = 4.2 \text{ K}$ placed directly below the sample. Measurements were done in the Faraday configuration in a superconducting solenoid. Both investigated samples were fully opaque in the energy range 85-220 meV due to phonon-related absorption in SiC.

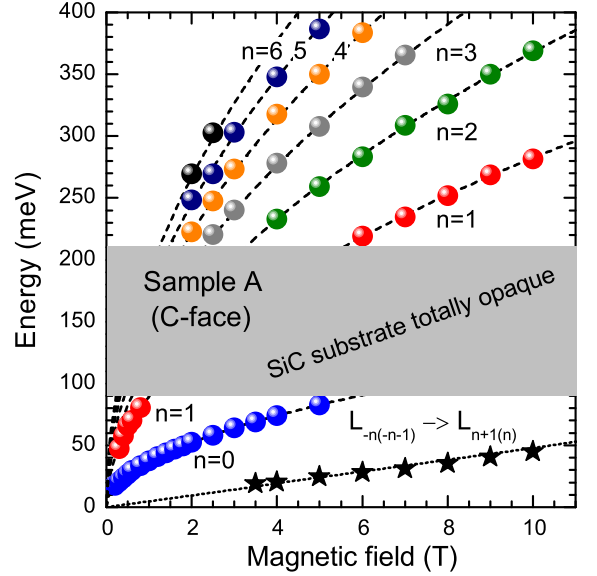


FIG. 2: (color online) Positions of absorption lines observed in the A sample grown on C-terminated surface of SiC. The spheres correspond to inter-LL transitions in electrically isolated and nearly undoped layers in MEG, having a characteristic \sqrt{B} -dependence. The dashed lines show their theoretically expected positions for the Fermi velocity of $v_F = 1.02 \times 10^6 \text{ m.s}^{-1}$. Origin of the transition denoted by stars, which evolves nearly linearly with B , is discussed in the text.

III. RESULTS AND DISCUSSION

Let us start our discussion with the data obtained on the sample A, i.e. on the MEG structure grown on the C-terminated surface of SiC, see Fig. 1, whose magneto-optical response is relatively well understood.^{10,13,22} Transmission spectra are in this kind of specimens dominated by relatively sharp absorption lines with a typical \sqrt{B} -like scaling (see Fig. 2) and taking the LL spectrum of graphene: $E_n = \text{sign}(n)v_F\sqrt{2\hbar|eBn|}$, we easily identify them as inter-LL transitions $L_{-n} \rightarrow L_{n+1}$ and $L_{-(n+1)} \rightarrow L_n$ with $n = 0 \dots 6$. The corresponding Fermi velocity can be evaluated as $v_F = (1.02 \pm 0.01) \times 10^6 \text{ m.s}^{-1}$. As a matter of fact, this response is equivalent to that of an exfoliated graphene monolayer^{11,12,23} and in turn it serves as a direct indication that layers in MEG indeed behave as independent, electrically isolated, graphene sheets.

Apparently, here we deal with graphene in its fully quantum regime, i.e. we study optical excitations between well-separated Landau levels, whose \sqrt{B} -scaling and multi-mode character are directly related to the linearity of the electronic bands.²⁴ To characterize our sample further, we can set the upper limit for the carrier density that cannot significantly exceed 10^{10} cm^{-2} , as the transition $L_{0(-1)} \rightarrow L_{1(0)}$ is observed down to the magnetic field of 100 mT (the filling factor $\nu < 6$). We assume that the graphene sheets are slightly p -doped as

follows from the recent ARPES study done on equivalent specimens.¹⁴ We should also note that the dominant graphene-like lines are often accompanied by additional spectral features, which are usually significantly weaker, nevertheless still well-defined in the spectra, located especially at low energies, see e.g. the transition denoted by stars in Fig. 1. We will come back to these features later on after we discuss the response of the sample B.

The sample B exhibits a very much different behavior in our magneto-transmission experiment and shows no \sqrt{B} -scaled spectral features, which are, as we have learned above, characteristic of graphene in its quantum regime. Instead, it clearly shows magnetic-field-induced transmission at low energies accompanied by a relatively wide absorption minimum, which moves towards higher energies with increasing B . We found this behavior to be characteristic for all graphene specimens prepared on Si-terminated surface of SiC (not shown in this paper), nevertheless, the total intensity of the observed spectral features as well as the position of the absorption minimum slightly varied from sample to sample.

To explain this behavior, we should recall that epitaxial graphene on the Si-terminated surface is usually highly doped by the charge transfer from the substrate, reaching carrier densities up to 10^{13} cm^{-2} ($E_F \approx 0.4 \text{ eV}$), see e.g. Ref. 20, with quality (expressed in terms of scattering time or mobility) comparable to MEG on the C-face of SiC.^{13,25} Therefore, at magnetic fields of a few Tesla, LLs should not be resolved around the Fermi level and the system should remain in the quasi-classical regime. Interestingly, owing to the non-equidistant spacing of LLs in graphene, the tuning from a fully quantum regime to the classical one is possible just by changing the Fermi level.

If we adopt this, the qualitative explanation of the observed behavior becomes straightforward. The relative magneto-transmission spectrum $T(B)/T(0)$ reflects two specific effects: The low-energy free carrier (Drude-like) absorption, which is gradually suppressed with the increasing field, and the cyclotron resonance itself. The former one gives rise to the field-induced transmission at low energies that decreases monotonically with the photon energy, and the latter one is manifested by an absorption minimum close to the energy of the cyclotron resonance ω_c . Let us now proceed with the quantitative analysis of this data, which will allow us to determine the characteristic field-dependence of ω_c .

In the framework of the classical theory for cyclotron resonance, the optical conductivity is given by

$$\sigma_{\pm}(\omega, B) = \sigma_0 \frac{i\gamma}{\omega \pm \omega_c + i\gamma}, \quad (1)$$

which describes both the CR active (−) and inactive (+) polarization modes of light, as well as the Drude-like low-energy absorption (at $B = 0$). σ_0 here denotes zero-field dc conductivity and γ is a phenomenologically introduced damping. We should also point out that in graphene, contrary to conventional systems of massive particles, the

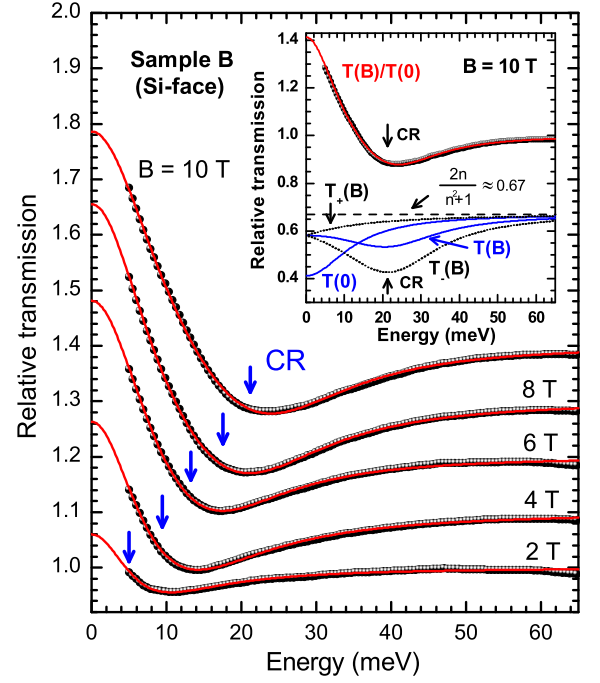


FIG. 3: (color online) Transmission spectra of the sample B taken at various magnetic fields in the far infrared spectral region. Solid curves represent fits to experimental data (spheres) according to the quasi-classical theory described in the text. For clarity, successive spectra are shifted vertically by 0.1. Vertical arrows indicate position of a relatively broad CR resonance, as obtained from the analysis. The inset shows fitting procedure at $B = 10 \text{ T}$ in detail. The individual transmission curves $T(0)$, $T(B)$, $T_+(B)$ and $T_-(B)$ have been plotted to demonstrate their contribution to the final change of sample transmission $T(B)/T(0)$. Let us note that these curves share a common baseline given by the transmission of the slab without any conducting layer (or away from the resonances) $T_{\text{slab}} = 2n/(n^2 + 1) \approx 0.67$.

cyclotron frequency ω_c is not independent of σ_0 and γ as the “effective” effective mass of Dirac fermions (at the Fermi level) can be determined from these two parameters.

The transmission of circularly-polarized light through the studied specimen (graphene sheet on an isolating substrate) can be expressed by formula:²⁶

$$T_{\pm}(\omega, B) = \frac{16n^2}{|\alpha_{\pm}(\omega, B)|^2 - |\beta_{\pm}(\omega, B)|^2}, \quad (2)$$

where

$$\alpha_{\pm}(\omega, B) = (n + 1) \left(n + 1 + \frac{\sigma_{\pm}(\omega, B)}{\varepsilon_0 c} \right)$$

$$\beta_{\pm}(\omega, B) = (n - 1) \left(n - 1 - \frac{\sigma_{\pm}(\omega, B)}{\varepsilon_0 c} \right)$$

which is derived for a dielectric slab with the index of refraction n ($n_{\text{SiC}} \cong 2.6$) with an infinitely thin conducting layer on one of its surfaces (described by σ_{\pm}). c

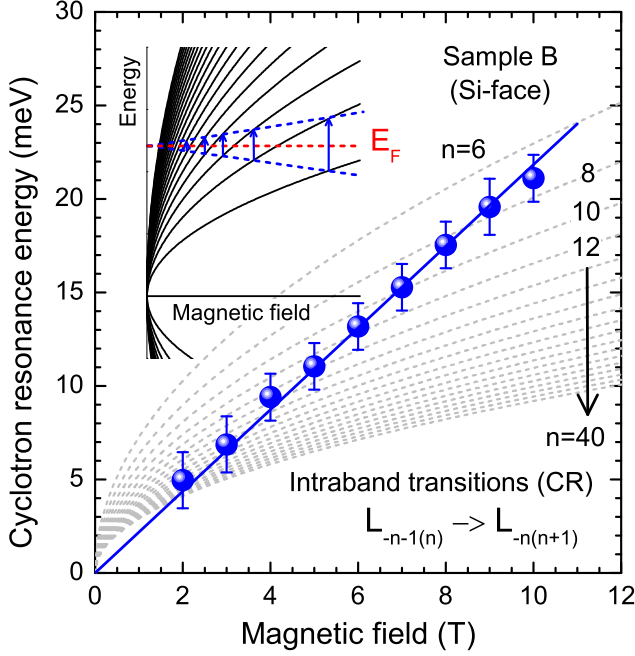


FIG. 4: (color online) Position of the cyclotron resonance in the sample B as extracted from the fitting experimental curves in Fig. 3, well-following the linear in B dependence plotted for the cyclotron mass of $m = 0.053m_0$ (full line). For comparison, we also depict positions of intraband (i.e. cyclotron) resonances between LLs with higher indices, $L_n \rightarrow L_{n+1}$ $n = 6 \dots 40$ (dotted lines). The inset schematically shows linear in B evolution of CR in highly doped graphene, despite the fact that the individual LLs exactly follow the \sqrt{B} -dependence. As we cannot determine the sign of charge of the particles in our non-polarized transmission experiment, we considered n -type doping only in this inset for simplicity.

and ε_0 stand for the real speed of light and the vacuum permittivity, respectively. The dependence of T_{\pm} on the thickness of the slab (i.e. interference effects) has been already averaged out.

Within the framework of our simple model, we can easily calculate the relative change of the sample transmission with the magnetic field $[T_+(B) + T_-(B)]/2T(0)$, and compare it with our experimental data. As the conductivity as well as the damping parameter can in principle vary with B , we have first found their appropriate values at zero field, $\sigma_0 = (2.4 \pm 0.3) \times 10^{-3} \Omega^{-1}$ and $\gamma_0 = (10 \pm 2) \text{ meV}$, which we afterwards kept constant and varied only ω_c , σ_B and γ_B to reproduce our experimental traces.

As shown in Figs. 3 and 4, our simple theoretical model is able to reproduce experimental transmission spectra very well and the CR frequency indeed follows a clear linear in B dependence. The conductivity σ_B and the CR half-width γ_B exhibited in our fitting procedure a weak decrease and increase, respectively, but practically remain within the interval of uncertainty estimated for σ_0 and γ_0 and they can be therefore taken as nearly constant in the investigated range of fields. The width (FWHM)

of the cyclotron resonance absorption peak $2\gamma_B$ has been found comparable with the cyclotron energy ω_c what justifies the quasi-classical regime in the investigated sample.

We should also clarify that a relatively broad CR resonance implies that the minimum in the transmission spectra does not exactly match to the position of the CR mode. This is explained in the inset of Fig. 3, where the transmission curves $T(B)$ at $B = 10 \text{ T}$ and $T(0)$ are depicted as calculated from parameters obtained in our data analysis. We also plot curves $T_-(B)$ and $T_+(B)$, which show expected transmission in opposite circular polarizations of light, i.e. in the CR active and inactive modes, respectively. Let us note that all these transmission curves share a common baseline at $2n/(n^2 + 1) \approx 0.67$, which corresponds to the light transmission through a dielectric slab without any conducting sheet (or away from resonances). This value can be easily obtained from Eq. (2) for $\sigma \equiv 0$.

Let us now return to the main finding of our analysis, i.e. to the clear linear in B dependence of the cyclotron resonance as shown in Fig. 4. If we evaluate the effective mass corresponding to this CR mode, we obtain $m = (0.053 \pm 0.004)m_0$, which according to the Einstein relation $E_F = mv_F^2$ implies the Fermi level of $E_F \approx 310 \text{ meV}$ in a reasonably good agreement with conclusions of ARPES measurements on similar samples.^{20,21} In this estimation, we used the Fermi velocity of $1.02 \times 10^6 \text{ m.s}^{-1}$ determined from the measurement on the sample A, but very similar velocities are reported by ARPES for graphene on the Si-terminated surface of SiC as well.^{20,21}

The observation of the CR mode that is linear in the applied field B justifies our simple approach in which (otherwise massless) Dirac fermions are considered as massive particles, the mass of which is determined by the Fermi energy. On the other hand, this confirmation implies another constrain on parameters used in our fitting. Namely, the conductivity σ_0 can be directly estimated within the Boltzmann transport theory from the scattering rate γ_0/τ and from the Fermi energy: $\sigma_0 = (2e^2/h)(E_F/\gamma_0)$. This relation was respected in our fitting procedure and required an additional self-consistent loop to keep the correct relation between σ_0 and γ_0 on the basis of E_F extracted from the fitting. Obviously, this approach works only in case of specimens with a full coverage by the graphene monolayer, otherwise the conductivity derived from γ_0 and E_F exceeds the value obtained directly from the fitting. The ratio of these two values thus could serve as a simple parameter that describes fraction of coverage by graphene.

We should note that the evaluated mass of $0.053m_0$ is surprisingly close the effective mass of K point electrons in bulk graphite $\sim 0.058m_0$,²⁷ but this similarity represents just a coincidence. To prove this, we can for instance argue that bulk graphite reaches the quantum limit at fields around 7 T ,^{28,29} and therefore it cannot exhibit CR line that corresponds to the effective mass

of K point electrons. Instead, due to the formal similarity of the band structure at the K point with the graphene bilayer, it should provide the CR-like absorption at energy by a factor of $\sqrt{2}$ higher, as indeed observed experimentally.³⁰

To summarize, the highly-doped graphene sheet on the Si-terminated surface allows us to study the CR resonance of massless Dirac fermions in its fully classical limit, when it becomes strictly linear with the applied magnetic field. Nevertheless, let us look at the same problem using the quantum-mechanical description, i.e. interpret our data as intra-band excitations between adjacent (but in reality significantly broadened) Landau levels, as schematically shown in the inset of Fig. 4. To clarify the linear in B response, found in a system with a \sqrt{B} -scaled LL spectrum, we only need to consider the Fermi level E_F located high in the conduction (or equivalently low in the valence) band and independent of the magnetic field. This can be exactly ensured by the pinning the Fermi level by the surface states in the substrate or fulfilled approximatively, when LLs significantly overlap. At a given magnetic field, the n -th LL approaches the Fermi level, $E_F = E_1\sqrt{n}$, and the active CR-like transition (between adjacent LLs) has the energy of $\hbar\omega_c = E_1(\sqrt{n+1} - \sqrt{n}) \approx E_1/(2\sqrt{n}) = \hbar eB/(E_F/v_F^2)$, i.e. it indeed becomes linear in B and moves with the slope related to the cyclotron mass $m = E_F/v_F^2$.

The analysis of data obtained on highly-doped graphene in the sample B allows us to come back to the low-energy features in spectra taken on the sample A. Also in this kind of samples, we usually find relatively weak spectral features whose energy moves nearly linearly in B . Even though the sample A dominantly consists of nearly undoped graphene sheets, the increase of the magneto-transmission at lower energies serves as an indication of some free carriers in the sample, similarly to the sample B. In fact, also in MEG on the C-face of SiC, first few layers nearby the interface are usually doped by charge transfer from the substrate. The transition denoted by stars (see Figs. 1 and 2) thus could be assigned to the CR response of these doped layers, and its characteristic slope $\approx 4.5 \text{ meV} \cdot \text{T}^{-1}$ (effective mass of $0.025m_0$) would imply the Fermi level of $|E_F| \approx 150 \text{ meV}$. On the other hand, the carrier density in these doped layers gradually decreases with the distance from the substrate, which should prevent observation of a well-defined CR mode (whose energy depends on the density of massless particles), while the transition denoted by the star remains relatively narrow in our data.

More likely, this line originates in Bernal-stacked residuals, especially in the graphene bilayer which has been

identified as a minor but not negligible component of MEG.^{31,35} The massive Dirac fermions in the bilayer are characterized by an effective mass of $m_B = \gamma_1/(2v_F^2) \approx 0.32m_0$ ($\gamma_1 = 380 \text{ meV}$ ³²⁻³⁵) which does not match to the effective mass derived for the “star” transition, but the characteristic LL spectrum in the bilayer, $E_n \cong \text{sign}(n)(\hbar|eB|/m_B)\sqrt{|n|(|n|+1)}$,^{36,37} implies in the quantum limit appearance of the CR mode ($L_{-1(0)} \rightarrow L_{0(1)}$ transitions) whose slope is by a factor of $\sqrt{2}$ higher. The transition denoted by the star, with a slope corresponding to the effective mass of $0.025m_0$, thus could be assigned an undoped graphene bilayer in which the CR absorption should correspond to the mass of $m_B/\sqrt{2} \approx 0.023m_0$. Nevertheless, the definitive conclusion cannot be drawn on the basis of our current data.

IV. CONCLUSIONS

We have explored magneto-optical response of graphene in two distinctively different regimes. Whereas the weakly doped sheets in multilayer epitaxial graphene on the C-terminated surface allowed us to study the optical response of graphene in the quantum limit, massless Dirac fermions in the quasi-classical regime can be explored in highly doped epitaxial graphene prepared on the Si-terminated surface of SiC. The response of the former system clearly shows \sqrt{B} -like scaling, typical of Landau levels in systems with linear electronic bands, the latter case proves that cyclotron resonance of massless particles remains in the quasi-classical limit linear with the magnetic field.

Additional note: During our work on the manuscript we became aware of Faraday rotation measurements on epitaxial graphene,³⁸ in which a similar conclusion about cyclotron resonance of massless Dirac fermions in the quasi-classical limit has been made.

Acknowledgments

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* Electronic address: milan.orlita@lncmi.cnrs.fr

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